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Validation of the MIMOSA-AURORA-IFDM model chain for policy support: Modeling concentrations of elemental carbon in Flanders

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ABSTRACT

The ability of a complex model chain to simulate elemental carbon (EC) concentrations was examined. The results of the model chain were compared to EC concentration measurements made at several locations, every sixth day. Two measurement campaigns were taken into account, one in 2006–2007 and one in 2008–2009. The model results compare very well for both periods, with an R^2 of 0.74, a bias of 0.02 μg m^{-3} and a RMSE of 0.32 μg m^{-3}. Sensitivity analyses to different meteorology inputs and changing emissions from year to year were performed. The differences between the two measurement periods were also investigated. It is shown that somewhat more than half of these differences is due to meteorology. However, emission changes also play an important role.

1. Introduction

Elemental carbon (EC) is considered to have a significant effect on human health (Dijkema et al., 2008; Patel et al., 2009; Lefebvre et al., 2011). It has also been found to be a suitable indicator of road traffic and it has been shown that exposure to abundant road traffic has significant impacts on several aspects of human health (Hoek et al., 2002; Gauderman et al., 2007; Mills et al., 2007). Therefore, the measurement and modeling of this parameter is gaining increasing attention. The Flemish Environment Agency (VMM) has set up two measurement campaigns at different locations in Flanders, in order to assess the chemical composition of PM_{10} (PM = particulate matter smaller than 10 μm), including EC and organic carbon (OC) concentrations. These campaigns were called Chemkar1 and Chemkar2. The first measurement campaign (Vercauteren et al., 2011), which was carried out at six different locations, was intended to assess the PM composition at various representative locations in Flanders. The second one was used to examine a number of hotspot locations for PM_{10} to obtain a better understanding of the reason for their hotspot-status.

The current European directive for air pollution (2008/50/EC) requires Member States to meet several air quality standards in every place within their territory. As it is impossible to measure at every location, it is strongly advised to combine air quality models with measurements at several well-chosen locations. This European guideline imposes limits on annual averaged concentration values or annual numbers of days/hours in exceedance of a threshold value. Therefore, it is necessary that a model is also validated on its capacity to simulate these annual statistics.

During the last decades, models have been developed and validated for several pollutants such as ozone or particulate matter. However, as science evolves, the focus changes and presently, it is aimed at more specific pollutants (e.g. PM_1, Ultrafine Particles (UFP) and EC). Therefore, as measurements are performed for these new pollutants, it is obvious that models should be tested and validated against these new datasets.

For the detailed simulation of air quality in larger domains, such as large urban areas or countries, a combination of several air quality models is required. The combination of models needs to be validated before it can be used in such an assessment. Since the seventies, validation and comparison studies have been performed on the models used in this study (e.g. Cosemans et al., 1981; Olesen, 1995; Thunis et al., 2009).

In this paper, the capability of the model chain, which consists of the traffic emission model MIMOSA, the Eulerian 3D grid dispersion

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model AURORA and the bi-Gaussian plume model IFDM, in simulating the EC concentrations is validated, not only at rural and urban background monitoring sites, but also at the more difficult hotspot locations. Furthermore, we try to assess the effect of the meteorology on the yearly mean concentrations. The same MIMOSA-AURORA-IFDM model chain is used for policy support in the framework of the EU Air Quality Directive.

The models have been applied to Flanders, the northernmost part of Belgium. It is a flat and densely populated area, with more than 6 million people residing on a surface area of more than 13,500 km^2, resulting in a population density of more than 450 persons km^{-2}. The area can be confined in a rectangle extending 238 km from west to east and 94 km from north to south.

2. Model description

2.1. Emissions

For the road traffic emissions, MIMOSA4 was used. MIMOSA4 is the most recent version of MIMOSA (Mensink et al., 2000; Vankerkom et al., 2009), which generates hourly output for different types of emissions, such as NO_2, PM_{10} and $PM_{2.5}$ for Flanders (see also Lefebvre et al., 2011). The latest version of MIMOSA4 relies on the COPERT 4 methodology (COPERT 4, 2007) for the energy consumption and emission functions for the conventional fuels (diesel, petrol and LPG). Next to this data, it is necessary to distribute the road network over the selected domain, together with the number and type of vehicles that use each road segment (De Vieger et al., 2011). The model not only aims at calculating total emission evaluations, but also calculates geographically distributed emissions over time. Indeed, the goal is not only to obtain the total emission over Flanders over a complete year, but to obtain emission data for each particular road segment for every hour.

The MIMOSA4 model had not yet the possibility to directly derive EC emissions. Therefore the model was extended for this new pollutant. The assumption was made that EC traffic emissions account for 66.11% of the $PM_{2.5}$ total traffic primary emissions. This fraction is based on the COPERT 4 emission factors for EC for the Flemish fleet composition. The high value can be explained by the large diesel fraction of the newly purchased vehicles over the last few years in Flanders (Lefebvre et al., 2011).

Although traffic emissions play a major role in this study, a complete emission inventory over all sectors is required for the dispersion model evaluation. Therefore, the non-traffic emissions of the different pollutants such as NO_x, PM_{10} and $PM_{2.5}$ for Flanders are based on the emission inventory compiled by the Flemish Environment Agency. For the other regions, the EMAP-tool (Maes et al., 2009) is used which provides gridded emissions based on the EMEP data set. The EC emissions are calculated based upon the $PM_{2.5}$-emissions per Selected Nomenclature for Air Pollution (SNAP) sector. The percentage of EC in $PM_{2.5}$ was taken from Schaap et al. (2004), except for road transport where 66.11% was used (see Table 1). In fact, Schaap et al. (2004) discusses BC and not EC. We have assumed that the EC fraction is equal to the BC fraction. This corresponds to the literature on this subject (Venkatachari et al., 2006; Hitzenberger et al., 2006; Quincey et al., 2009), which estimates the BC/EC regression slope consistently in the immediate neighborhood of 1.

Table 1
The ratio $EC/PM_{2.5}$ as used in this study for the different SNAP-sectors, based on Schaap et al. (2004) except for S7. The last column is the estimated Flemish emission of EC per snap-sector based on the latest estimations of the $PM_{2.5}$-emissions from the Flemish Environment Agency (VMM) for 2007.

Number	Description	$EC/PM_{2.5}$	EC (ton yr^{-1})
S1	Combustion in energy and transformation industries (stationary sources)	0.11	88
S2	Non-industrial combustion plants (stationary sources)	0.21	498
S3	Combustion in manufacturing industry (stationary sources)	0.25	404
S4	Production processes (stationary sources)	0	0
S5	Extraction and distribution of fossil fuels and geothermal energy	0.85	0
S6	Solvent use and other product use	0	0
S7	Road transport	0.6611	2213
S8	Other mobile sources and machinery	0.52	1634
S9	Waste treatment and disposal	0.004	1
S10	Agriculture	0.17	85
S11	Other sources and sinks	0	0

2.2. AURORA

The atmospheric dispersion model used for simulation of the regional air quality in this study is 'Air quality modeling in Urban Regions using an Optimal Resolution Approach' (AURORA, Mensink et al., 2001). The AURORA model uses the method of nested simulations. In this model, the vertical diffusion is calculated with the Crank-Nicholson method (De Ridder and Mensink, 2002), while the horizontal advection uses a Walcek (2000) scheme. The gas phase chemistry is treated by the Carbon-Bond IV scheme (Gery et al., 1989), which has been enhanced to take into account biogenic isoprene emissions. For particulate matter (PM_{10} and $PM_{2.5}$), a distinction was made between primary and secondary particles. Both the amount and distribution of green vegetation cover are based on SPOT-VEGETATION satellite imagery. Terrain height is taken from the Global 30 Arc-second Elevation Data Set, distributed by the U.S. Geological Survey. Meteorological fields, required as input for AURORA, were simulated using the Advanced Regional Prediction System (ARPS) model, a non-hydrostatic mesoscale atmospheric model developed by the University of Oklahoma (Xue et al., 2000, 2001). More information on the AURORA model can be found in the European Model Database (http://air-climate.eionet.europa.eu/databases/MDS/index_html).

A validation of the combined ARPS-AURORA model setup can be found in De Ridder et al. (2008) and in Van de Vel et al. (2010) for the Ruhr area and the Lake Baikal region respectively. The model has also been validated for Flanders in several projects. The AURORA model has been used to simulate, with a high spatial resolution of 1×1 km^2, air pollution over Belgium (including Flanders) and for several European cities such as Rotterdam and Prague as part of the GMES-PROMOTE project. In this project, it has been demonstrated (PROMOTE, 2009a,b) that the AURORA model is able to accurately simulate air quality in urban regions. Analyses of the air quality over the Po-Valley in Italy for the POMI project show that the AURORA model performance is in general in line with respect to other similar high resolution models (Thunis et al., 2009). This model is also validated for the city of Shenyang, China (Lefebvre et al., 2010). Finally, the coupling of AURORA with both the MIMOSA emission model and an activity-based traffic model is validated in Beckx et al. (2009).

2.3. IFDM

The Immission Frequency Distribution Model (IFDM) model is a bi-Gaussian plume model, designed to simulate non-reactive pollutant dispersion on a local scale. The Gaussian dispersion parameters are dependent on the stability of the atmosphere and the wind speed following the Bultynck and Malet formulation

based on the Bulk Richardson number (Bultynck and Malet, 1972). Line sources are treated as in Venkatram and Horst (2006), except for the cases where the wind is parallel or almost parallel to the road. In the latter case, numerical integration of a series of point sources is applied. Area sources are treated as a set of equivalent parallel line sources perpendicular to the wind. Currently, the IFDM model only uses meteorology from one fixed point for the complete domain. The model will be adapted in the future, in order to use spatially differentiated meteorology measurement data. As IFDM is a receptor-model, it can be used for every grid setup, whether it is regular or not. More information on the IFDM model can be found in the European Model Database (http://air-climate.eionet.europa.eu/databases/MDS/index_html).

2.4. AURORA-IFDM coupling

As mentioned in the introduction, air quality models can be used as tools for policy support for the new EU Air Quality Directive. However, in such a framework air pollution has to be simulated over a large area such as the Flemish region (13,522 km^2) with sufficient detail to account for the large gradients along highways and major point sources. Therefore, it is insufficient to rely only on a 1–3 km scale 3D grid model such as AURORA, as its results are not at a high enough resolution or a 25 m–1 km scale bi-Gaussian plume model such as IFDM. In order to combine the best of both worlds, the two models are coupled to cover both the regional aspects of the air pollution phenomenon and the large gradients along the major line and point sources.

One major problem that has to be accounted for in the coupling procedure is the double counting of emission sources. For example: when the result of the bi-Gaussian plume model is simply superimposed on top of the 3×3 km^2 grid concentration of the regional air quality model, all emissions taken into account by the plume model are counted twice. In our approach, the AURORA and IFDM model are coupled by using a simple algorithm to avoid double counting of the sources:

1. First of all, AURORA simulates air pollution on a resolution of $3 \times 3km^2$, using all sources (case A in Fig. 1).
2. Secondly, IFDM simulates on a regular grid (finer than AURORA, 488×194 points on a 0.48×0.48 km^2 resolution, 39 IFDM-grid points per AURORA-cell) the air pollution from (traffic) sources which are of interest (in this case, traffic emissions on the major roads) (case B in Fig. 1).
3. Thirdly, the concentration in each AURORA-cell is adapted cell by cell, by subtracting from its hourly concentrations, the spatial mean of the hourly concentrations of all IFDM receptor points in this cell (case C in Fig. 1). This results in AURORA-concentrations without the effect of the local traffic sources which are of interest (case D in Fig. 1). These AURORA concentrations are interpolated on the IFDM-grid (see below), using a bi-linear interpolation technique (case E in Fig. 1).
4. Finally, IFDM simulates the air pollution due to the traffic sources which are of interest (same sources as in the second point). In this step, the model uses a road following grid (ranging from 1000×1000 m^2 away from the roads till

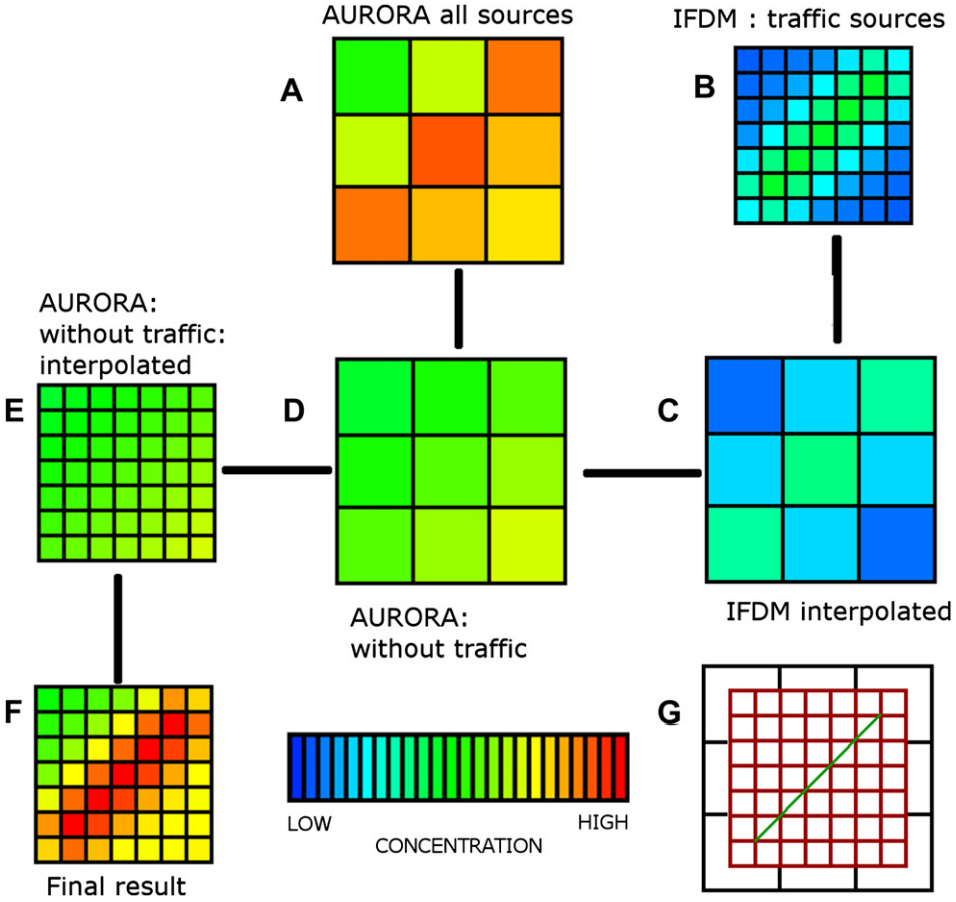

Fig. 1. The different steps in the coupling of AURORA and IFDM. Case G denotes the different grids: in black AURORA, in red IFDM. In this example, in order to keep the figure simple, only a regular IFDM grid has been shown. The green line is a road as the major emission source in this example. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

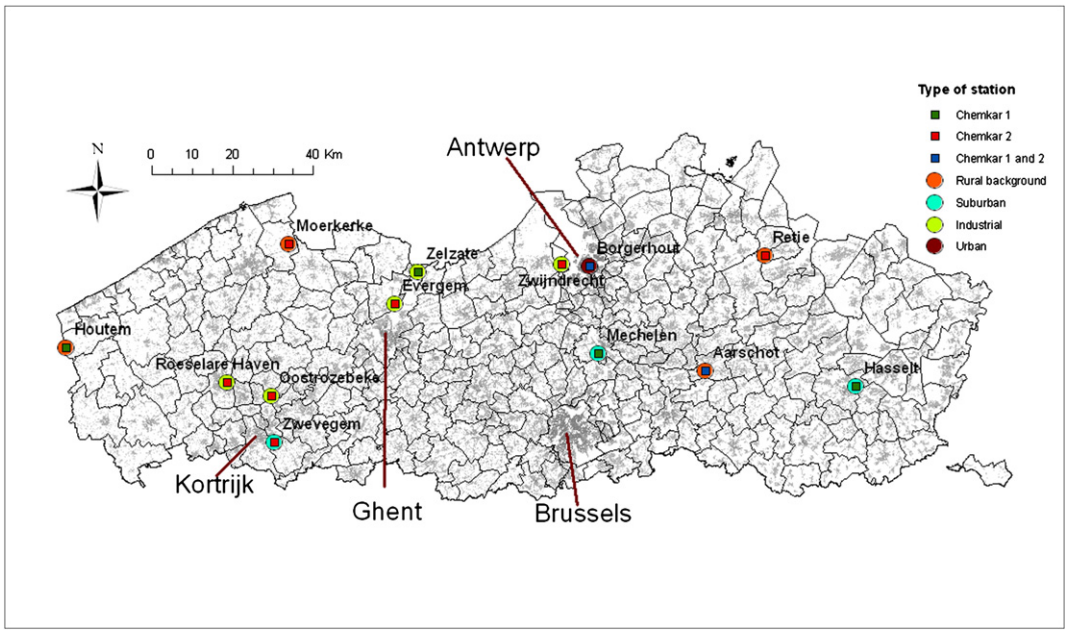

Fig. 2. The locations of the Chemkar monitoring sites, superimposed on a population map of Flanders and Brussels (gray = densely populated). In circles, the type of monitoring site is shown (e.g. rural background, suburban, …). The squares within the circles show the Chemkar-campaign in which the monitoring site was included.

$25 \times 600\ m^2$ close to the roads following a similar methodology as in Lefebvre et al. (2011)), in order to have more receptor points available where the largest gradients are expected. These IFDM-results are added to the concentrations calculated in point 3. As a result, a detailed hourly concentration field is created with a regional pattern and steep gradients along the major line (and point, if applicable) sources (case F in Fig. 1), without double counting.

The AURORA-IFDM coupling procedure is similar to the one used in Lefebvre et al. (2011).

3. Measurement campaign

3.1. Sampling sites

For Chemkar1, sampling was done at six sites in Flanders, Belgium (Fig. 2) from September 2006 to September 2007. The sites were chosen to obtain a reasonable spread in location and type of site. The six sites were located in Houtem (rural background), Zelzate (industrial), Mechelen (suburban), Borgerhout (urban), Aarschot (rural background) and Hasselt (suburban). For Chemkar2 sampling was done at nine sites in Flanders, Belgium, between

Table 2
Characteristics of the different simulations used in this study. The resolutions 'IFDM regular' and 'IFDM road following' are described in §3.4. The "mean conc" column denotes the modeled mean concentration (in $\mu g\ m^{-3}$) averaged over all monitoring sites available in at least one measurement campaign (and counting Aarschot and Borgerhout only once).

Simulation	Resolution	Model	Emissions	Emission year	Meteorology	Mean conc ($\mu g\ m^{-3}$)
AUR07	$3 \times 3\ km^2$	AURORA	All	2007	Modeled 2007	1.03
AUR10	$3 \times 3\ km^2$	AURORA	All	2010	Modeled 2007	0.91
I07_ARPS_r	IFDM regular	IFDM	Flemish Roads	2007	Modeled 2007	0.26
I10_ARPS_r	IFDM regular	IFDM	Flemish Roads	2010	Modeled 2007	0.21
I07_ARPS	IFDM road following	IFDM	Flemish Roads	2007	Modeled 2007	0.34
I10_ARPS	IFDM road following	IFDM	Flemish Roads	2010	Modeled 2007	0.28
I_06_07	IFDM road following	IFDM	Flemish Roads	2007	Measured 2006	0.32
I_07_07	IFDM road following	IFDM	Flemish Roads	2007	Measured 2007	0.30
I_08_07	IFDM road following	IFDM	Flemish Roads	2007	Measured 2008	0.31
I_09_07	IFDM road following	IFDM	Flemish Roads	2007	Measured 2009	0.27
I_CK1_07	IFDM road following	IFDM	Flemish Roads	2007	Measured Chemkar1	0.34
I_CK1_10	IFDM road following	IFDM	Flemish Roads	2010	Measured Chemkar1	0.28
I_CK2_07	IFDM road following	IFDM	Flemish Roads	2007	Measured Chemkar2	0.29
I_CK2_10	IFDM road following	IFDM	Flemish Roads	2010	Measured Chemkar2	0.23
BACK07	$3 \times 3\ km^2$	AUR07-I07_ARPS_r				0.77
BACK10	$3 \times 3\ km^2$	AUR10-I10_ARPS_r				0.70
CK1_comp	IFDM road following	BACK07 + CK1_07				1.11
CK2_comp	IFDM road following	1/3*(BACK07 + CK2_07) + 2/3*(BACK10 + CK2_10)				0.97
CK2-CK1	IFDM road following	CK1_comp-CK2_comp				−0.14
ΔE_b	IFDM road following	2/3*(BACK10 − BACK07)				−0.05
ΔE_l	IFDM road following	2/3*(CK2_10 − CK2_07)				−0.03
ΔM_l	IFDM road following	CK2_07 − CK1_07				−0.06

October 2008 and November 2009. Six of the sites were considered PM_{10} hot spots: Borgerhout (same site as in Chemkar1), Roeselare, Oostrozebeke, Zwevegem, Evergem and Zwijndrecht; three were considered rural background sites: Aarschot (same as in Chemkar1), Moerkerke and Retie.

3.2. Sampling and EC/OC analysis

For a detailed description of the Chemkar approach we refer to Vercauteren et al. (2011). In short, 24-h sampling was carried out simultaneously on all locations on every sixth day with a PM_{10} low-volume sampler (Leckel SEQ 47/50). With this apparatus 55 m^3 of air was sampled on a 47 mm diameter quartz fiber filter (pre-fired Whatman QM-A). After weighing, the filters were cut for different types of analysis. For EC/OC determination a 1 or 1.5 cm^2 punch was analyzed by means of a thermal–optical transmission (TOT) method with a Sunset Laboratory (Tigard, OR, USA) Lab OCEC analyzer using the NIOSH 5040 protocol (Birch, 2003). This method is known to give relatively low EC values compared to some other techniques and protocols, but at the same time the NIOSH-EC is probably the best indicator for traffic emissions.

3.3. Meteorology measurements

Where measured meteorology has been used in this study, it is taken from the Luchtbal pylon in Antwerp. At this urban location, temperature, wind speed and direction are measured at a height of 30 m.

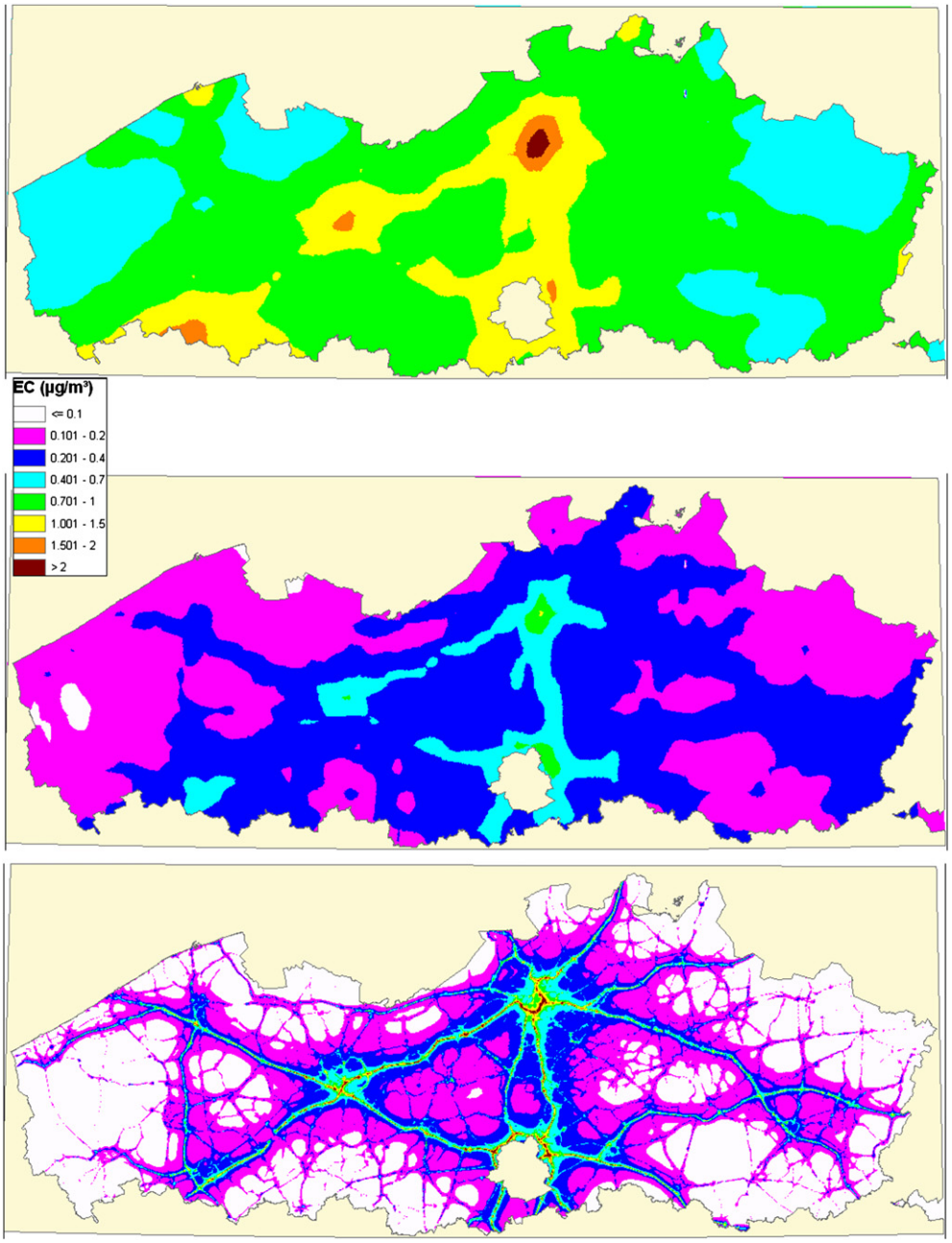

Fig. 3. The different components of the EC concentration (in µg m^{-3}). Top: the EC concentration as simulated in *AUR07*. Middle: the concentration as simulated in *I07_ARPS_r* averaged out over 3 × 3 km^2. Bottom: the concentration as simulated in *I_CK1_07*, the road contribution. The final simulation *CK1_comp* is calculated as the upper panel minus the middle panel plus the lower panel and is shown in the top part of Fig. 4.

4. Model setup and simulations

Up until now, a very detailed hourly by hourly simulation methodology has been described. However, as EC is a passive pollutant and as we estimate that the background emissions do not show much inter-annual variability, we can use the annual mean background calculations for the different years, all made with 2007 meteorology.

To validate the coupled model chain with the measurements collected during the two Chemkar campaigns, several simulations with the different models were made (Table 2):

- AURORA simulations, nested in 60 × 60 km^2 BelEUROS-output (Deutsch et al., 2008a,b, 2009) at resolutions of respectively 25 × 25 km^2, 9 × 9 km^2 and 3 × 3 km^2 for the years 2007 and 2010. As the simulations were made in 2010, i.e. before emission and meteorology data for 2010 were available, emission projections by the Flemish Environmental Agency in cooperation with VITO have been used. Furthermore, meteorology from 2007 was used for both simulations and is calculated by ARPS based on the ECMWF-reanalysis for the year 2007. Thus, differences between 2007 and 2010 are due to changes in emissions. The simulations at 3 × 3 km^2 are named *AUR07* and *AUR10*.
- IFDM traffic EC simulations for the years 2007 and 2010 using the ARPS-meteorology for 2007. Only Flemish traffic emissions derived by the MIMOSA4 model are taken into account. These simulations are named *I07_ARPS_r* and *I10_ARPS_r* on the regular IFDM-grid and *I07_ARPS* and *I10_ARPS* on the road-following grid.
- IFDM traffic EC simulations using measured meteorology data from the years 2006, 2007, 2008 and 2009, with EC emissions from the year 2007. Only Flemish traffic emissions are taken into account. These simulations are named *I_06_07*, *I_07_07*, *I_08_07*, *I_09_07* respectively.
- IFDM traffic EC simulations using measured meteorology data for the collection dates of the measurement campaigns with EC emissions both for the year 2007 and for the year 2010. These simulations are named I_*CK1_07*, I_*CK2_07*, I_*CK1_10* and I_*CK2_10* respectively. Only Flemish traffic emissions are taken into account.

Using the results from the first two bullet points, background concentrations without the roads for the year 2007 and 2010 can be calculated (steps A–C, Fig. 1) in the AURORA-IFDM coupling. The results are called *BACK07* and *BACK10*. Thereafter, depending on the situation, one of the IFDM simulations can be used to create the final concentration set.

The Chemkar measurement campaigns, Chemkar1 and Chemkar2, were conducted mostly during the year 2007, and the year 2009 respectively. Therefore, in order to compare the simulated results with the results of the Chemkar1-campaign, only emission data for 2007 were used (*BACK07* + *I_CK1_07*, we call this the *CK1_comp*). For the Chemkar2-campaign however, a blend of both 2007 and 2010 data were used (1/3*(*BACK07* + *I_CK2_07*) + 2/3*(*BACK10* + *I_CK2_10*), we call this the *CK2_comp*). These are the datasets that will be validated and discussed in this paper.

For the location of the cities mentioned in this chapter, we refer to Fig. 2. The different contributions to the overall *CK1_comp* simulation are presented in Fig. 3. In the top panel, the AURORA-results are shown. Increased concentrations are observed around the major highways and in the major cities (especially around Ghent, Antwerp and Brussels). However, another area with high EC concentration is apparent in the southwestern part of Flanders. On

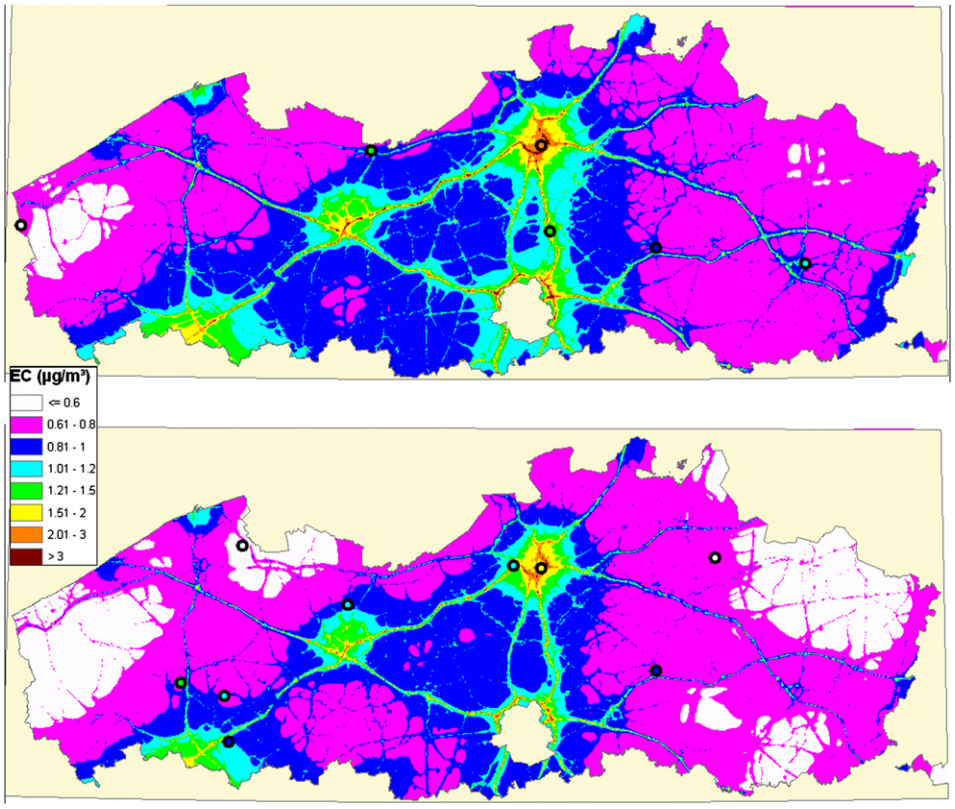

Fig. 4. The modeled EC concentration (in µg m^{-3}) in the *CK1_comp* simulation (top) and in the *CK2_comp* simulation (bottom). The circles are the monitoring sites and are filled corresponding to the measured concentrations.

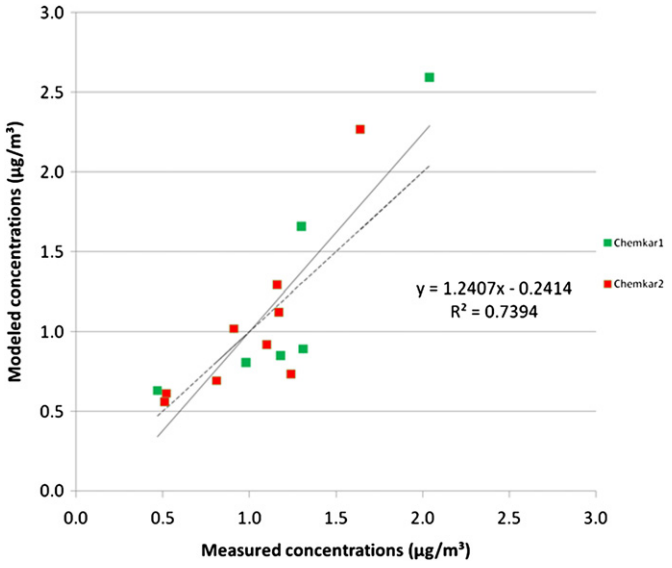

Fig. 5. The validation graph for the campaign-mean EC concentrations. On the *X*-axis: the measured EC concentrations in µg m^{-3}. On the *Y*-axis: the modeled EC concentrations in µg m^{-3}, using *CK1_comp* results for Chemkar1-monitoring sites and *CK2_comp* results for Chemkar2-monitoring sites. Every dot represents one monitoring site during one campaign: green for Chemkar1, red for Chemkar2. The dashed line is the 1–1 line. The black full line shows the regression between the measurements and the model, for which the equation and the R^2 are shown on the right side. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

analyzing the AURORA results, it is shown to be related to a significant import of EC from the highly urbanized region around Lille in northern France.

In the middle panel of Fig. 3, the effect of the Flemish road emissions, smoothed out over a 3 × 3 km^2 grid is shown. As can be observed, these emissions are mainly located around the urban city centers of Ghent, Brussels, Antwerp and Kortrijk and around some major highways (especially, the highways Kortrijk–Ghent–Antwerp, Ghent–Brussels and Brussels–Antwerp). As explained before, the simulation *BACK07* is created by subtracting these results from the *AUR07*-simulation. The high resolution impact of the roads can be examined in the bottom panel of Fig. 3. In this figure all major roads are clearly visible. It is clear that large gradients exist in the immediate neighborhood of these traffic sources. At a certain distance away from these roads the EC concentrations are low. However, close to the important traffic emission sources, the EC concentrations are very high with a maximum of about 9 µg m^{-3} on the Ring of Antwerp. The resulting simulation, *CK1_comp* is shown in the top panel of Fig. 4. As expected high concentrations are revealed around the major highways and in the city centers of Antwerp and Ghent. Furthermore, high concentrations can be found at the Flanders-France border south of Kortrijk.

5. Results

5.1. Comparison to measurements

In order to compare the results of the MIMOSA-AURORA-IFDM model chain to measurements, measurements of the Chemkar1-campaign are compared with the *CK1_comp*-dataset and the measurements of the Chemkar2-campaign with the *CK2_comp*-dataset, using the average over the complete time series per station.

As can be seen in Fig. 5, the results of the comparison are very promising. The R^2 value is 0.739, while the bias (about 0.02 µg m^{-3}) and the RMSE (about 0.32 µg m^{-3}) are small. Furthermore, the regression line (model ~ = 1.2407*measurement−0.2414) is not too far from the 1:1-line. Apart from this comparison, the different parts of the simulation chain can also be evaluated. This evaluation is summarized in Table 3. Herein the combined version performs best with a high R^2, a very small bias and a low RMSE. However, the different components do not perform too badly either. For instance, the AURORA simulations themselves (including traffic) show an even lower RMSE, although the R^2 is somewhat lower. The bias is also low in this case. The Flemish traffic simulations only resulted in an R^2 close to 0.7. However, the bias and as a result also the RMSE are much higher. This is logical, as only part of the existing emissions are used in these simulations. Finally, the simulations without the Flemish traffic emissions also exhibit a larger bias and a larger RMSE combined with a lower R^2. This again can be explained as part of the emissions is also missing.

Table 3
Comparison of the different simulations with the measurements. The measurement values are collected in column "Meas". The "Combined" column combines the "CK1_comp" and the "CK2_comp" column by taking the value of the CK1_comp column if the measurement is performed during the Chemkar1-campaign and of the CK2_comp column otherwise. All model and measurement values are in µg m^{-3}. The mean value is based on the averaged value of all data, counting both the Aarschot and the Borgerhout data for both years (and thus double for the model simulations).

Nr	Cp	Location	Meas	AUR07	AUR10	BACK07	BACK10	I_CK1_07	I_CK2_07	I_CK2_10	CK1_comp	CK2_comp	Combined
1	CK1	Houtem	0.47	0.6450	0.5430	0.6228	0.5325	0.0080	0.0120	0.0090	0.6308	0.5726	0.6308
2	CK2	Moerkerke	0.51	0.6270	0.5540	0.5605	0.5035	0.0520	0.0430	0.0350	0.6125	0.5602	0.5602
3	CK2	Retie	0.52	0.7050	0.5970	0.6216	0.5273	0.0690	0.0590	0.0510	0.6906	0.6124	0.6124
4	CK1	Aarschot	0.98	0.8420	0.7250	0.5999	0.5233	0.2070	0.1600	0.1360	0.8069	0.6928	0.8069
	CK2	Aarschot	0.81	0.8420	0.7250	0.5999	0.5233	0.2070	0.1600	0.1360	0.8069	0.6928	0.6928
5	CK2	Zwevegem	0.91	1.1580	0.9680	1.0739	0.9050	0.0740	0.0650	0.0530	1.1479	1.0183	1.0183
6	CK2	Evergem	1.10	0.9780	0.9010	0.8121	0.7607	0.2000	0.1530	0.1360	1.0121	0.9195	0.9195
7	CK2	Zwijndrecht	1.16	1.4290	1.3110	0.8599	0.8497	0.5270	0.5000	0.4090	1.3869	1.2924	1.2924
8	CK2	Oostrozebeke	1.17	0.8900	0.7760	0.7585	0.6695	0.6100	0.4600	0.4060	1.3685	1.1232	1.1232
9	CK1	Hasselt	1.18	0.8550	0.7390	0.6382	0.5543	0.2130	0.1590	0.1340	0.8512	0.7246	0.8512
10	CK2	Roeselare H	1.24	0.8690	0.7680	0.6774	0.6129	0.1270	0.1130	0.0930	0.8044	0.7341	0.7341
11	CK1	Mechelen	1.30	1.2020	1.0490	0.8354	0.7491	0.8240	0.6240	0.5290	1.6594	1.3385	1.6594
12	CK1	Zelzate	1.31	0.8530	0.7630	0.6735	0.6134	0.2180	0.1680	0.1470	0.8915	0.7874	0.8915
13	CK1	Borgerhout	2.04	2.3030	2.1560	1.2673	1.2988	1.3270	1.1100	0.9120	2.5943	2.2663	2.5943
	CK2	Borgerhout	1.64	2.3030	2.1560	1.2673	1.2988	1.3270	1.1100	0.9120	2.5943	2.2663	2.2663
		R^2	1.000	0.675	0.680	0.543	0.611	0.696	0.692	0.694	0.703	0.698	0.739
		Mean (µg m^{-3})	1.09	1.10	0.98	0.79	0.73	0.40	0.33	0.27	1.19	1.04	1.11
		Bias (µg m^{-3})	0.00	0.01	−0.11	−0.30	−0.36	−0.69	−0.76	−0.82	0.10	−0.05	0.02
		RMSE (µg m^{-3})	0.00	0.29	0.30	0.41	0.45	0.73	0.80	0.85	0.37	0.30	0.32

5.2. Estimation of the EC carbon concentrations due to Flemish road emissions

Based on these analyses (§2.6 and 4.1), it can be concluded that, on average[1] about 25–31% of the EC concentrations can be attributed to Flemish traffic (Table 2, see also lower panel Fig. 3, and upper panel Fig. 4). This number can be calculated by, for instance for the year 2007, comparing the mean of the I_CK1_07 simulation to the mean of the CK1-comp simulation (0.40/1.11 = 31%). The remaining part is due to Flemish non-traffic emissions or to non-Flemish (both traffic and non-traffic) emissions. The proportion due to Flemish traffic emissions varies from about 1% in Houtem to about 50% in Mechelen and Borgerhout (Table 3). The percentage due to Flemish traffic is slightly higher when the 2007 emissions are used instead of the 2010 emissions. This indicates that in this period EC traffic emissions are decreasing faster than other sector contributions, thanks to renewal of the car technology and despite the increasing use of diesel cars during this period.

5.3. Further sensitivity tests

The results of further sensitivity tests can be found in the Supplementary material.

6. Discussion and conclusions

Based on the results of the comparison (with the measurements) presented in this paper, we can conclude that the MIMOSA-AURORA-IFDM model chain presented here is well capable of simulating the spatial variability of EC concentrations averaged over a longer period. Furthermore, it is shown that the meteorology can play an important role in the EC concentrations. However, when annual mean concentrations are considered, the meteorology effects are reduced. Further sensitivity tests have been made and were discussed in the Supplementary material. Finally, it can be concluded that on the basis of these results, the currently used $EC/PM_{2.5}$ ratio in the Flemish road emissions inventory yields good results.

The EC concentrations in Flanders are shown to be between 0.6 and 1 $\mu g\ m^{-3}$ in rural areas away from major roads (Fig. 4) and between 1 and 3 $\mu g\ m^{-3}$ in urban areas, excluding street canyons. These figures are comparable to the ones found in the Supplementary material of Putaud et al. (2010) for the rural case in Northwestern Europe and lower than the values in Central Europe. However, the methodology used to determine EC is different from study to study and the difference in the results can reach as high as 100% (Putaud et al., 2010). We know that the methodology used in our study results in low EC concentrations and therefore, it is not possible to claim a significant difference between our results and those in the rest of Europe. Furthermore, as we have shown, EC concentrations are decreasing in time, at least in Flanders. Therefore, it can be problematic to compare EC concentrations from studies ranging over two decades.

The maps resulting from this effort can be used by the Flemish government and other interested parties in determining the possible risks of the population to the exposure to elemental carbon. Furthermore, this model chain can be used in order to assess elemental carbon concentrations at locations where no measurements are available. As measurements of elemental carbon are expensive, this model chain can provide a relative cheap way in determining which locations risk being exposed to high elemental carbon concentrations. Furthermore, it can assist the responsible agency to determine suitable measurement locations for elemental carbon.

[1] averaged over all monitoring sites in the Chemkar dataset and depending on the simulation.

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Appendix. Supplementary material

Supplementary data related to this article can be found online at doi:10.1016/j.atmosenv.2011.08.033.

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